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## Measurement of Ferroelectric Liquid Crystal Parameters

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A method is presented for measuring ferroelectric liquid crystal parameters such as spontaneous polarization  $P_s$ , rotational viscosity  $\gamma_\phi$ , azimuthal pretilt angle  $\phi_0$  and dielectric permittivity  $\epsilon_\perp$ . The method is based on integrating the polarization current after reversing the sign of the external electric field.

### INTRODUCTION

Ferroelectric liquid crystals (FLC), discovered by Meyer in 1975<sup>1</sup>, have become a subject for intensive research and have found increasing application as electrooptical media for displays and data processing devices. At present, they are successfully used in fast-response light shutters, image converters, alpha-numeric data displays and flat television screens.

The major physical parameters which define the speed of response for FLC electrooptical devices are the spontaneous polarization  $P_s$ , and the rotational viscosity  $\gamma_\phi$ . There are three basic approaches to measuring the spontaneous polarization: the pyroelectric method<sup>2</sup>, the Sawyer-Tower procedure<sup>3</sup>, and a set of methods based on measurements of the repolarization current of a liquid crystal cell<sup>4</sup>. The first of these approaches offers the highest accuracy for measurement of  $P_s(T)$ , especially for low values of spontaneous polarization, but it is complicated to implement and measuring is time-consuming. The second approach is rapid but allows only the  $P_s$  parameter to be measured. The third group of methods involves measuring the area under the curve for the repolarization current and thus requires additional time to be spent on processing of experimental results.

Rotational viscosity is measured using the current repolarization method<sup>4</sup> and electrooptical procedure<sup>7</sup>. Thus, to measure the magnitude of spontaneous polarization and rotational viscosity, it is necessary to use several different methods.

The authors propose a universal procedure for measuring FLC parameters which enables the values of  $P_s$  and  $\gamma_\phi$  to be obtained from a single experiment. In addition, this experiment gives the magnitudes of the azimuthal pretilt angle  $\phi_0$  and the dielectric permittivity  $\epsilon_\perp$ .

### EXPERIMENTAL PROCEDURE AND THEORETICAL BACKGROUND

A schematic diagram of the system used for measuring physical parameters of an FLC is shown in Figure 1. Current ( $i$ ) passing through an FLC cell is integrated by a capacitor  $C^*$  connected in series with the cell and a voltage generator. During measurement of the spontaneous polarization  $P_s$ , the voltage form of the generator can be arbitrary; however, alternating voltage of a rectangular form seems to be the most convenient because both the spontaneous polarization and the rotational viscosity can then be measured in the same experiment. The schematic diagram of Figure 1 is, in fact, one arm of the capacitance bridge in the Sawyer-Tower circuitry. In the present case, however, the X-axis is scanned over the time rather than the voltage enabling us to measure the time dependence of the voltage across  $C^*$ — $U_{out}(t)$ —which varies due to integrating the current which is passing through the FLC cell.

Let us assume that a ferroelectric liquid crystal is oriented in such a way that smectic layers are perpendicular to electrodes (Figure 2). When a voltage  $U$  is applied to the electrodes of the cell, the vector of spontaneous polarization  $\mathbf{P}$ , tends to lie in the direction of the electric field  $\mathbf{E}$ , while the azimuthal angle  $\varphi$  between them tends to zero. In the process of reorientation of  $\mathbf{P}$ , there occurs a repolarization current  $i_p$  associated with the change in the surface charge on the electrodes. In addition, through the cell there passes a current  $i_r$  (see inset in Figure 1) which is associated with the ionic conduction of the liquid crystal. The third component is the displacement current,  $i_c$ , related to cell capacity recharging consequently, the instantaneous value for the total current through the cell can be presented as the sum of the three terms:

$$i = i_r + i_c + i_p = \frac{U}{R} + C \cdot \frac{dU}{dt} + S \cdot \frac{dP_s}{dt} \quad (1)$$

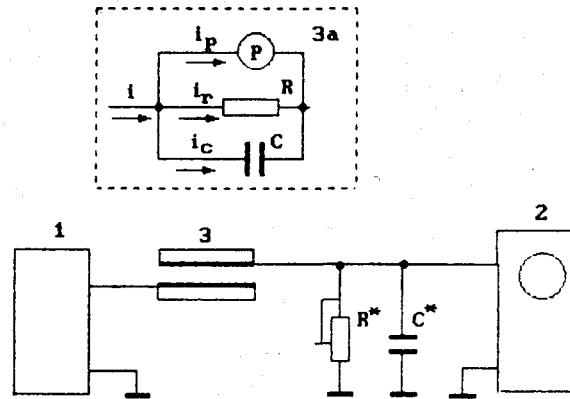


FIGURE 1 Schematic diagram of the system: 1—functional generator, 2—oscilloscope, 3—LC cell. Inset 3a shows the equivalent electric diagram of the LC cell:  $C$  is capacity of the cell,  $R$  is resistance of the cell,  $P$  is a nonlinear element associated with the spatial orientation of the spontaneous polarization vector  $\mathbf{P}_s$ .

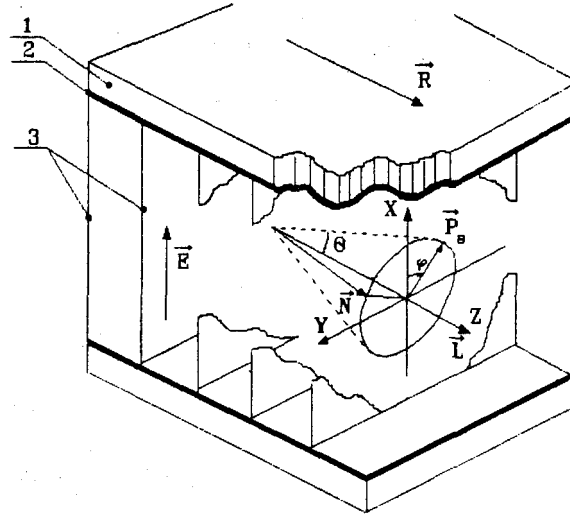


FIGURE 2 FLC cell structure and coordinate system: 1—quartz glass, 2—transparent  $\text{SnO}_2$  electrode, 3—smectic layers;  $R$ —direction of rubbing the orienting layer,  $E$ —electric field vector,  $L$ —normal to the smectic layers,  $P_s$ —spontaneous polarization vector,  $N$ —FLC director,  $\theta$ —angle of the molecular tilt,  $\phi$ —azimuthal angle, X, Y, Z—axes of the coordinate system of the cell.

where  $C = (\epsilon_{\perp} \cdot \epsilon_0 \cdot S)/d$  is the capacity of the cell, and  $d$ ,  $S$  and  $R$  are the thickness, area and resistance of the cell, respectively, and  $U$  is voltage across the cell. It is noted that, in the general case, the time dependence of the cell resistance is sophisticated but here we assume  $R$  is constant, which is true for short  $t$ . The instantaneous value for the voltage across the capacitor  $C^*$  will be:

$$U_{out} = -\frac{1}{C^*} \int i dt = \frac{U}{RC^*} + \frac{2UC}{C^*} + \frac{P_s S}{C^*} = U_r + U_c + U_p \quad (2)$$

where  $t=0$  represents the moment of reversing the sign of the electric field across the cell.

Figure 3 shows oscillograms of the driving voltage from the function generator and the output voltages across the capacitor  $C^*$ . Section 1 corresponds to the voltage  $U_p$ . The voltage generator time constant is much shorter than the oscilloscope sweep time and, for this reason, the oscillogram shows it as an instantaneous jump. That is followed by smooth growth in the voltage (section 2) due to integration of the re-polarization current  $i_p$  and the conductivity current  $i_r$ . Section 3 is due to the ionic conduction current  $i_r$ . In practice,  $i_p \gg i_r$ , and section 2 can be ascribed to the integration of the repolarization current only. Otherwise, the effect of  $i_r$  can be eliminated by introducing a variable resistance  $R^*$  into the measuring circuit (Figure 1).

By measuring the corresponding values  $U_p$  and  $U_c$  (Figure 3) and using the formulas

$$P_s = \frac{U_p C^*}{2S} \quad (3a)$$

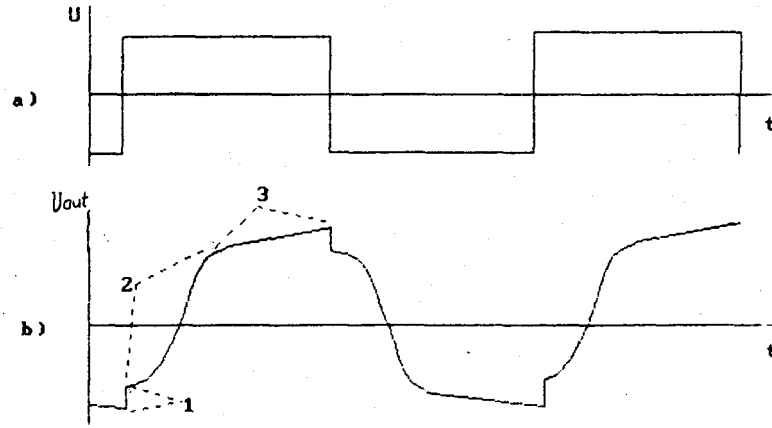


FIGURE 3 Oscillograms for (a) the driving voltage and (b) output signal from the capacitor  $C^*$ .

and

$$\varepsilon_{\perp} = \frac{U_c d C^*}{2 \varepsilon_0 S U} \quad (3b)$$

one can find the spontaneous polarization  $P_s$  and the dielectric constant  $\varepsilon_{\perp}$ . We will now show how rotational viscosity  $\gamma_{\varphi}$  can be determined using measurements of time dependence  $U_p(t)$ . In the general case, the equation for the angle dynamics  $\varphi(t)$  governed by rotational viscosity  $\gamma_{\varphi}$  (in the one-constant approximation) can be written as

$$\gamma_{\varphi} = P_s E \cdot \sin \varphi + \varepsilon_0 \Delta \varepsilon \cdot E^2 \cdot \sin^2 \theta \cdot \sin \varphi \cdot \cos \varphi + K \cdot \nabla^2 \varphi \quad (4)$$

where  $\theta$  is the angle of the molecular tilt in the FLC layer and  $K$  is the elastic constant. For sufficiently large fields<sup>5</sup>,  $E > (\Delta \gamma)^2 / P_s K$  with  $\Delta \gamma$  being a coefficient of surface interaction, a simultaneous conical rotation of molecules occurs in the smectic layer with the cone apex  $2\theta$ , and Equation 5 is reduced to<sup>6</sup>

$$\gamma_{\varphi} = P_s E \cdot \sin \varphi + \varepsilon_0 \Delta \varepsilon \cdot E^2 \cdot \sin^2 \theta \cdot \sin \varphi \cdot \cos \varphi \quad (5)$$

The exact solution of this equation is as follows<sup>6</sup>:

$$\frac{t}{\tau} = \frac{1}{1 - \alpha^2} \cdot \left\{ \ln \frac{\operatorname{tg}(\varphi / 2)}{\operatorname{tg}(\varphi_0 / 2)} + \ln \frac{(1 + \alpha \cdot \cos \varphi) \cdot \sin \varphi_0}{(1 + \alpha \cdot \cos \varphi_0) \cdot \sin \varphi} \right\} \quad (6)$$

where  $\tau = \gamma_{\varphi} / P_s E$ ,  $\varphi_0 = \varphi(t = 0)$ ,  $\alpha = \varepsilon_0 \Delta \varepsilon \cdot E \cdot \sin^2 \theta / P_s$ . Having found  $\varphi(t)$  from Equation 6, one can find the repolarization current and voltage over the capacitor  $C^*$  as a function of  $t$ :

$$U_p = \frac{S P_s}{C^*} \cdot \cos \varphi(t) \quad (7)$$

$$i_p(t) = S \frac{d(P_s \cos \varphi(t))}{dt} = \frac{SP_s}{\tau} \sin \varphi \cdot (1 + \alpha \cos \varphi(t)) \quad (8)$$

Let us consider a case with  $\alpha = 0$ . Then Equation 5 has a solution in the explicit form<sup>7</sup>:

$$\varphi(t) = 2 \arctg(f), \quad (9)$$

where  $f = tg(\varphi_0/2) \exp(t/\tau)$ . In this case expressions for  $U_p$  and  $i_p$  are simpler:

$$U_p(t) = \frac{2SP_s}{C^*} \cdot \frac{f^2}{1+f^2} \quad (10)$$

and

$$i_p(t) = 4SP_s \cdot \frac{f^2}{(1+f^2)^2} \quad (11)$$

From Equation 10 it follows that

$$t_{10-90} = \ln(9) \cdot \gamma_\varphi / P_s E \approx 2.2 \cdot \gamma_\varphi / P_s E \quad (12)$$

where  $t_{10-90}$  is rise time of the voltage  $U_p$  from 10% to 90% of the maximum level (Figure 3). Similarly, for other time intervals we get

$$t_{20-80} = \ln(4) \cdot \gamma_\varphi / P_s E \approx 1.4 \cdot \gamma_\varphi / P_s E \quad (13a)$$

$$t_{30-70} = \ln(7/3) \cdot \gamma_\varphi / P_s E \approx 0.85 \cdot \gamma_\varphi / P_s E \quad (13b)$$

$$t_{40-60} = \ln(3/2) \cdot \gamma_\varphi / P_s E \approx 0.4 \cdot \gamma_\varphi / P_s E \quad (13c)$$

The switching times  $t_{10-90}$ ,  $t_{20-80}$ , and  $t_{30-70}$  as a function of the parameter  $\alpha$  are given in Figure 4. It can be seen that in the range  $|\alpha| < 0.4$  the relative deviation of  $t_{10-90}$  from the value for  $|\alpha| = 0$  does not exceed 5%. For the time interval  $t_{20-80}$  this range expands up to  $|\alpha| < 0.6$ , and for  $t_{30-70}$   $|\alpha| < 0.95$ . The interval  $t_{40-60}$  is

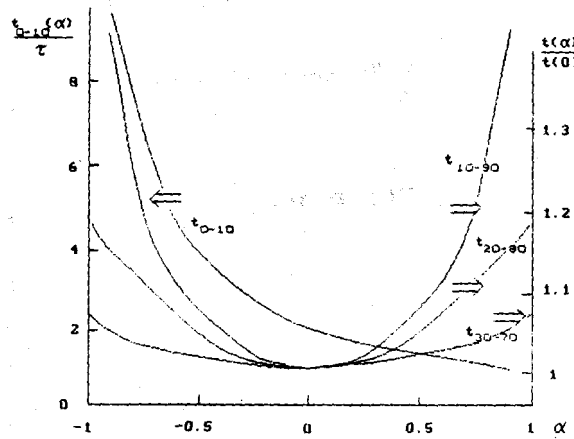
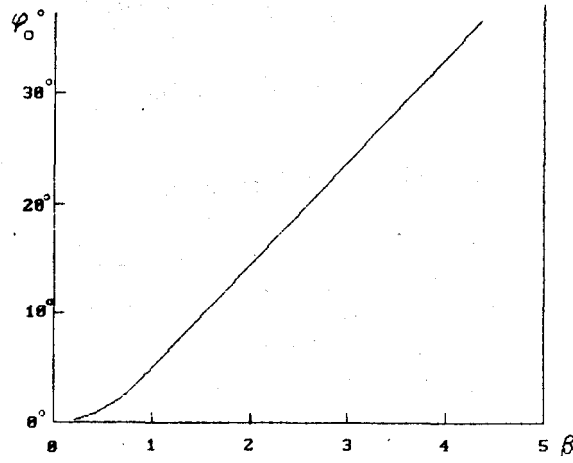


FIGURE 4 Calculated plots of time intervals versus parameter  $\alpha$ .

FIGURE 5 Calculated plots of azimuthal angle  $\varphi_0$  versus parameter  $\beta$ .

practically independent of  $\alpha$ . Consequently, in calculating  $\gamma_\varphi$  shorter intervals of the function  $U_p(t)$  should be used.

Of great interest is the evaluation of the initial azimuthal angle  $\varphi_0$ , as this defines to a significant degree the switching delay time  $t_{10-90}$ . Solving Equation 11 for  $\varphi_0$  we get

$$\varphi_0 = 2 \cdot \arctg\left(\frac{1}{3 \exp(\ln 9 / \beta)}\right) \quad (14)$$

where  $\beta = t_{10-90}/t_{0-10}$ . This function is plotted in Figure 5. It should be noted that  $t_{0-10}$  depends significantly on  $\alpha$  (see Figure 4); that is why Equation 14 can be used for evaluation of  $\varphi_0$  only for the range  $|\alpha| < 0.05$ .

It is noted also that Equation 3 for  $P_s$  is true only for the case  $\varphi_0 = 0$ , so in the general case ( $\varphi_0 \neq 0$ ) the measured value  $P_s^*$  is a function of  $\varphi_0$ .

$$P_s^* = P_s^* \cdot \cos \varphi_0 \quad (15)$$

However, for  $\varphi_0 < 20^\circ$ , applicable in practice, the error for the measurement of  $P_s$  does not exceed 5%.

## RESULTS OF FLC PARAMETER MEASUREMENTS

To verify the proposed procedure, comparative measurements were made of some FLC parameters. The experiments were carried out on the ferroelectric liquid crystalline material ZhKS-52 composed of nonchiral smectic C and an optically active dipole dopant. Its ferroelectric phase temperature interval ranged from  $+2^\circ\text{C}$  to  $+50^\circ\text{C}$ .

The LC cell consisted of two parallel quartz plates with transparent electrodes on their inner surfaces. The gap between the glasses (from 2 to 15  $\mu\text{m}$ ) was fixed by

Teflon spacers. Planar orientation of the liquid crystal was achieved through thin layers of PVA applied onto the electrodes. The layers were rubbed in the same direction on both glasses. The FLC cell was filled at the temperature corresponding to its isotropic phase.

Figure 6 shows the results of the temperature measurements of spontaneous polarization  $P_s$  and rotational viscosity  $\gamma_\phi$  made using the procedure proposed here, compared with measurements of  $P_s$ , made using the pyroelectric method<sup>2</sup> and of  $\gamma_\phi$  made by the electrooptic route<sup>7</sup>. The rotational viscosity  $\gamma_\phi$  was calculated from the dynamic  $U_p$  curves using Equation 12 and from the electrooptical measurements using the following formula<sup>6</sup>:

$$\gamma_\phi = t_{10-90}^* P_s E / 1.75 \quad (16)$$

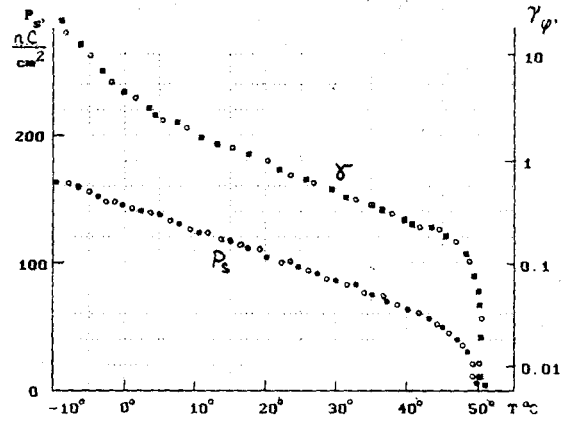


FIGURE 6 Temperature dependences of spontaneous polarization and rotational viscosity. Key: ● pyroelectric method for  $P_s$  ■ electro-optical method for  $\gamma_\phi$ ; ○ procedure proposed in this paper for both  $P_s$  and  $\gamma_\phi$

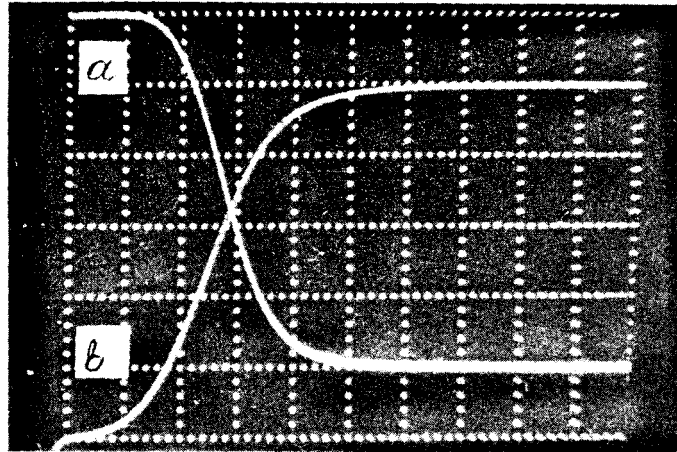


FIGURE 7 Oscillograms for (a) electro-optical response and (b) output signal from the capacitor  $C^*$ .



Oscillograms of the time dependences  $U_p$  and optical transmission are presented in Figure 7. From the oscillograms we get the relationship  $t_{10-90}/t_{10-90}^* \approx 1.3$  which coincides to a high degree of precision with the magnitude of this ratio obtained from Equations 12 and <sup>16</sup>:

$$t_{10-90}^* / t_{10-90}^* = 2.2 / 1.75 \approx 1.26$$

The values of spontaneous polarization obtained via the different procedures differ by not more than by 5%, while those of rotational viscosity by less than 10%. The original azimuthal angle  $\varphi_0$  calculated for our cells from Equation 14 was  $12^\circ$ , while that obtained from the optical measurements according to the procedure given in Reference 8 was  $10^\circ$ .

## CONCLUSIONS

The procedure proposed for measuring parameters of ferroelectric liquid crystals, based on measurements of the repolarization current, allows one to estimate the spontaneous polarization  $P_s$ , rotational viscosity  $\gamma_\varphi$ , initial azimuthal angle at the cell walls  $\varphi_0$  and dielectric permittivity  $\varepsilon_\perp$ . The results obtained for  $P_s$ ,  $\varphi_0$  and  $\gamma_\varphi$  are in good agreement with measurements obtained using pyroelectric ( $P_s$ ) and electrooptical procedures ( $\gamma_\varphi$ ,  $\varphi_0$ ). The advantage of the proposed method is that the parameters are determined in a single experiment, from the relaxation voltage curves corresponding to the repolarization current of a liquid crystal cell. In addition, the measurements of  $P_s$  and  $\gamma_\varphi$  in this procedure are (in contrast to the electro-optical method) independent of the quality of the LC orientation and layer thickness.

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